Two approaches simulating Brownian motion in fluid suspensions(Poster session 1, New Frontiers in Colloidal Physics : A Bridge between Micro- and Macroscopic Concepts in Soft Matter)

AUTHOR(S):

Nakayama, Yasuya; Iwashita, Takuya; Yamamoto, Ryoichi

Two approaches simulating Brownian motion in fluid suspensions

Yasuya Nakayama[a1] , Takuya Iwashita[b] , Ryoichi Yamamoto[b,c]

[a]Depertment of Chemical Engineering, Kyushu University

[b]Depertment of Chemical Engineering, Kyoto University

[c]CREST, Japan Science and Technology Agency

コロイド粒子などのブラウン運動は，現象論的にはブラウン粒子についてのマルコフなランジュバン方程式でよく理解されている．よって、もっとも簡便なブラウン運動のシミュレーションは、ブラウン粒子だけで閉じたランジュバン方程式を解くことでなされる．しかし、ブラウン粒子だけで閉じた式において溶媒を介する流体力学相互作用を考慮することは容易ではない．これは，流体力学相互作用を考慮するために溶媒自由度を解くモデル化シミュレーションや直接シミュレーション法が多く提案されている理由の１つである．ブラウン粒子と溶媒自由度による系についてランジュバン方程式を導くと、熱ゆらぎは溶媒におけるランダム応力で表わされる，そのため溶媒自由度に対してゆらぎを加えることは１つの有効な直接シミュレーション法を与える．一方で、多くの場合に興味あるのは，ブラウン粒子の運動のみであることを考えると，圧倒的に多い溶媒自由度へのランダム力を解くことは効率的でない．さらに溶媒がニュートン流体以外の場合には，ランダム力同定のための摩擦テンソルのモデル化や導出さえも困難となる．我々は，揺動散逸定理もしくは Onsager's regression hypothesis に基づいて，効率的かつ拡張の容易な流体力学相互作用を考慮したブラウン運動のシミュレーション法を提案する．

Dynamics of sub-micron/nanoscale objects offers many intriguing physcial problems since both hydrodynamic interactions and thermal fluctuation are equally influence the motion of such objects. Among such objects, colloids and/or macromolecules have been typical targets of research. We propose a simple and applicable way for simulating colloidal suspensions incorporating hydrodynamic interactions and thermal fluctuation.

Before introducing our new scheme, we observe what we already know in the set of basic equations to describe the Brownian motion in fluid suspensions [1] [2] , viz.,

$$
\begin{aligned}
&\nabla \cdot \boldsymbol{v} = 0, \quad \rho \dot{\boldsymbol{v}} = \nabla \cdot \boldsymbol{\sigma} + \nabla \cdot \boldsymbol{s}, \\
&M\dot{\boldsymbol{V}}_i = \boldsymbol{F}_i + \boldsymbol{F}'_i, \quad \boldsymbol{I} \cdot \dot{\boldsymbol{\Omega}}_i = \boldsymbol{N}_i + \boldsymbol{N}'_i, \\
&\boldsymbol{F}_i = \int \mathrm{d}\boldsymbol{S}_i \cdot \boldsymbol{\sigma}, \quad \boldsymbol{F}'_i = \int \mathrm{d}\boldsymbol{S}_i \cdot \boldsymbol{s}, \\
&\boldsymbol{N}_i = \int (\boldsymbol{x} - \boldsymbol{R}_n) \times \mathrm{d}\boldsymbol{S}_i \cdot \boldsymbol{\sigma}, \quad \boldsymbol{N}'_i = \int (\boldsymbol{x} - \boldsymbol{R}_n) \times \mathrm{d}\boldsymbol{S}_i \cdot \boldsymbol{s},
\end{aligned}
\tag{1}
$$

[1]E-mail: ynakayama-w@chem-eng.kyushu-u.ac.jp

where $\rho, \boldsymbol{v}$ are the mass density and the velocity of the host fluid, respectively. $M, \boldsymbol{I}, \boldsymbol{R}_i, \boldsymbol{V}_i, \boldsymbol{\Omega}_i$ are the mass, the moment of inertia, the center of mass, velocity, and angular velocity of the ith colloid, respectively. Thermal fluctuation of the system is incorporated through the random stress $\boldsymbol{s}$ which is white-in-time and white-in-space,

$$\langle \boldsymbol{s}(\boldsymbol{x}, t)\boldsymbol{s}(\boldsymbol{x}', t')\rangle = 2k_B T\boldsymbol{\zeta}\delta(\boldsymbol{x} - \boldsymbol{x}')\,\delta(t - t'), \tag{2}$$

where the fourth rank friction tensor $\boldsymbol{\zeta}$ should be identified form the stucture of the viscous stress $\boldsymbol{\sigma}$. Equations (1) and (2) consists a large Markovian Langevin system. Therefore, not only Brownian motion of colloids but that of fluid particles is solved by this set of equations.

We propose another set of working equations by focusing to solve the Brownian motion solely of what we are interested in, colloids and/or macromolecules. Fluctuation–Dissipation theorem (FDT) on colloids states that velocity correlation of hot system ($k_B T > 0$) is equal to the velocity response function of the cold system ($k_B T = 0$). Thus, we omit the random stress in the equation of the host fluid but retain the viscous stress which is necessary for solving the velocity response. In the hot system, random stress $\boldsymbol{s}$ plays a role in sustaining the temperature of the whole system, but has no coherent effect on average regression of the velocity correlation of colloids since $\boldsymbol{s}$ is white-in-time and white-in-space. To agitate colloids, we need to impose random forces on colloids. These new random forces should be white-in-time in order not to have no coherent effect on the regression of the velocity correlation of colloids. The discussion above leads our set of working equations that is described Eq.(1) with,

$$\begin{aligned} \boldsymbol{s} &= \boldsymbol{0}, \\ \boldsymbol{F}'_i &= \boldsymbol{G}^V_i, \quad \langle \boldsymbol{G}^V_i(t)\boldsymbol{G}^V_i(0)\rangle \propto \delta(t), \\ \boldsymbol{N}'_i &= \boldsymbol{G}^\Omega_i, \quad \langle \boldsymbol{G}^\Omega_i(t)\boldsymbol{G}^\Omega_i(0)\rangle \propto \delta(t), \end{aligned} \tag{3}$$

Since FDT was not applied to the whole system, the amplitudes of the fluctuating forces $\boldsymbol{G}^V_i$ and $\boldsymbol{G}^\Omega_i$ need to be determined implicity. This is done by controlling $\langle \boldsymbol{V}^2\rangle(t)$, $\langle \boldsymbol{\Omega}^2\rangle(t)$ to certain target values.

By considering the FDT or Onsager's regression hypothesis on colloids, we derived a new set of equations for simulating Brownian motion in fluid suspensions. We conclude the presentation by making two remarks. Our scheme is highly efficient since simulation with Eqs.(1) and (3) requires much less degrees of freedom for random forces than Eqs.(1) and (2). Moreover, we note that our scheme is highly applicable to various type of host fluids since Eqs.(1) and (3) do not depend on a specific constitutive equation of the host fluid. Numerical results based on our scheme are reported elsewhere [3].

References

[1] E. H. Hauge and A. Martin-Löf: J. Stat. Phys. **7** (1973) 259.

[2] E. J. Hinch: J. Fluid Mech. **72** (1975) 499.

[3] T. Iwashita, Y. Nakayama and R. Yamamoto: to be published.